

## PATENT COOPERATION TREATY

PCT

## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Assistant Commissioner for Patents  
United States Patent and Trademark  
Office  
Box PCT  
Washington, D.C.20231  
ETATS-UNIS D'AMERIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 17 July 2000 (17.07.00)	
International application No. PCT/NL99/00676	Applicant's or agent's file reference P22013PC00
International filing date (day/month/year) 04 November 1999 (04.11.99)	Priority date (day/month/year) 04 November 1998 (04.11.98)
Applicant BAYENSE, Cornelis, Roeland et al	

1. The designated Office is hereby notified of its election made:



in the demand filed with the International Preliminary Examining Authority on:

31 May 2000 (31.05.00)



in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland  Facsimile No.: (41-22) 740.14.35	Authorized officer  Olivia RANAIVOJAONA  Telephone No.: (41-22) 338.83.38
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order to minimise pressure drop, the use of star shaped extrudates would be most suitable. However, star shaped bodies, extrudates, tend to be prone to attrition due to the presence of the 'points' of the star.

5           It is an object of the invention to reconcile these various requirements in the form of a transition alumina extrudate, having a carefully balanced set of properties. Further objects and advantages will become clear from the following description of the invention and the preferred  
10   embodiments thereof.

          The invention is based thereon that the inventors have now been able to provide a star shaped alumina extrudate, having on the one hand an optimal structure, as indicated above and on the other hand a good strength.

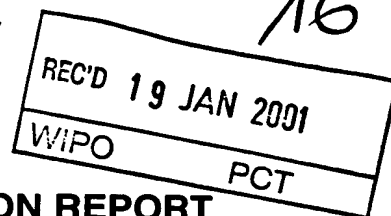
15           The invention is accordingly directed to star shaped alumina extrudates with a pore volume in pores of diameter of over 1000 nm, as determined by mercury porosimetry, of at least 0.05 ml/g, a side crushing strength of at least 50 N and a bulk crushing strength of at least 1 MPa.

20           Surprisingly, this set of properties can be made available in one material, thereby providing a material with which chemical reactions can be made much more efficient, resulting in higher activity and/or selectivity. Also the material of the invention, when used in fixed bed reactors,  
25   provides a decreased pressure drop compared to regular extrudates having a cylindrical shape.

          The BET surface area, as determined by single point adsorption using the BET equation (as e.g. described by G. Sandstede et.al., Chem. Ing. Tech. 32 (1960), 413), should be  
30   at least 10 m<sup>2</sup>/g of alumina. This coincides with the requirement of using a transition alumina, i.e. not an  $\alpha$  alumina. Suitable alumina's are the various transition alumina's including  $\gamma$ -alumina,  $\delta$ -alumina,  $\epsilon$ -alumina,  $\kappa$ -alumina,  $\zeta$ -alumina,  $\theta$ -alumina and  $\tau$ -alumina. These alumina's  
35   have a large BET-surface area, generally in the range of 25 up to more than 100 m<sup>2</sup>/g.

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)



Applicant's or agent's file reference P22013PC00	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/NL99/00676	International filing date (day/month/year) 04/11/1999	Priority date (day/month/year) 04/11/1998
International Patent Classification (IPC) or national classification and IPC B01J35/02		
Applicant ENGELHARD CORPORATION, ET AL		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.



2. This REPORT consists of a total of 6 sheets, including this cover sheet.

- ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 1 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand  31/05/2000	Date of completion of this report  17.01.2001
Name and mailing address of the international preliminary examining authority:   European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer  de Cauwer, R  Telephone No. +49 89 2399 7344  

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/NL99/00676

**I. Basis of the report**

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments (Rules 70.16 and 70.17).)*:

**Description, pages:**

1,3-6	as originally filed	
2	with telefax of	24/11/2000

**Claims, No.:**

1-9	as originally filed
-----	---------------------

**Drawings, sheets:**

1/1	as originally filed
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2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/NL99/00676

- ☐ the description, pages:  
☐ the claims, Nos.:  
☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

## V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

### 1. Statement

Novelty (N)	Yes:	Claims	1-9
	No:	Claims	
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1-9
Industrial applicability (IA)	Yes:	Claims	1-9
	No:	Claims	

2. Citations and explanations  
**see separate sheet**

## VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:  
**see separate sheet**

**Re Item V**

**Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**Claim 1 - Novelty**

Claim 1 is a product claim for star shaped alumina extrudates with a pore volume of at least 0.05 ml/g in the pores with a diameter of over 1000 nm, a side crushing strength of at least 50 N and a bulk crushing strength of at least 1 MPa.

EP-A-0 008 424 (BASF AG) 5 March 1980 (1980-03-05), D1, is regarded as being the closest prior art to the subject-matter of claim 1 and discloses (the references in parentheses applying to this document): star shaped extrudates (page 5, line 26-29) made from  $\text{Al}_2\text{O}_3$  (page 5, line 24) having a pore volume between 0.4 and 0.8 ml/g (page 5, line 5) with a high mechanical stability (page 4, line 22) and a pressure resistance of 10 kg (page 6, line 20).

The subject-matter of claim 1 therefore differs from that known in D1 in that the catalyst has a pore volume of at least 0.05 ml/g in the pores with a diameter of over 1000 nm. Although D1 states that the majority of the pores should be between 5 and 20 nm, this is obviously an error since this implies that the pores should be larger than the particles (examples page 6).

The subject-matter of claim 1 is therefore novel (Article 33 (2) PCT).

**Claim 1 - Inventive step**

The problem to be solved by the present invention is to provide a product that has a high mechanical strength and to eliminate diffusion limitation problems as much as possible.

The solution to this problem proposed in the present application is to provide a product with a pore volume of at least 0.05 ml/g in the pores with a diameter of over 1000 nm. This solution cannot be considered as involving an inventive step (Article 33 (3) PCT) since D1 solves the same technical problems as the current application, notably the provision of a product with a good mechanical stability (page 4, line 23) and a high activity, which obviously only can be achieved if there are no real problems of diffusion limitation. Furthermore no evidence has been given that the current product has any

advantage over the product of D1. Moreover, it is not unreasonable to assume that the pore volume in pores of diameter of over 1000 nm in the product of D1 is at least 0.05 ml/g, since this is only about 10% of the total pore volume of the product of D1 as well as in the current product. As a result it cannot be seen how the current product may be advantageous over the product of D1 and no inventive step can be recognised.

#### Claim 2 - 6 - Novelty & Inventive Step

Since the subject-matter of claim 1 is novel, that of the dependent claims 2 - 6 is novel too (Article 33 (2) PCT).

With regard to the inventive step, the same reasoning as for claim 1 applies and thus, the subject-matter of claims 2 - 6 is not inventive (Article 33 (3) PCT).

#### Claim 7 - Novelty & Inventive Step

Claim 7 is a product claim for a catalyst comprising at least one catalytically active material supported on an extrudate according to claims 1 - 6.

D1 discloses that the extrudate can be impregnated with a cobalt-nitrate solution (page 5, line 34).

Since the product of claim 1 is novel, the product of claim 7 can be regarded as novel too (Article 33 (2) PCT).

Since the problem to be solved is the same as for claim 1, the same reasoning applies as for claim 1, and thus, the subject-matter of claim 7 is not inventive (Article 33 (3) PCT).

#### Claim 8 - Novelty & Inventive Step

D1 further discloses that the catalytically active material can be cobalt (page 5, line 34). For these reasons and because claim 8 is dependent on claim 7, the subject-matter of claim 8 can be regarded as novel but not inventive (Article 33 (2) & (3) PCT).

#### Claim 9 - Novelty & Inventive Step

Claim 9 is a use claim of the products of claim 1 to 8 in a chemical reaction. The use of a novel product has to be considered novel too (Article 33 (2) PCT).

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/NL99/00676

D1 discloses the use of the catalyst for hydrogenation reactions (claim 1, line 2).  
Thus, the subject-matter of claim 9 cannot be considered inventive (Article 33 (3) PCT).

**Re Item VII**

**Certain defects in the international application**

To meet the requirements of Rule 5.1 a) ii) PCT, the document D1 should be identified in the description and the **correct** relevant background art disclosed therein should be briefly discussed.

**Re Item VIII**

**Certain observations on the international application**

The subject-matter of claim 9 is considered unclear (Article 6 PCT) since it refers to the use of two different products.



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

<b>(51) International Patent Classification <sup>7</sup> :</b> <b>B01J 35/02, 21/04</b>	<b>A1</b>	<b>(11) International Publication Number:</b> <b>WO 00/25918</b> <b>(43) International Publication Date:</b> 11 May 2000 (11.05.00)
<b>(21) International Application Number:</b> PCT/NL99/00676 <b>(22) International Filing Date:</b> 4 November 1999 (04.11.99)  <b>(30) Priority Data:</b> 98203719.4 4 November 1998 (04.11.98) EP  <b>(71) Applicant (for all designated States except US):</b> ENGELHARD CORPORATION [US/US]; 101 Wood Avenue, Iselin, NJ 08830-0770 (US).  <b>(72) Inventors; and</b> <b>(75) Inventors/Applicants (for US only):</b> BAYENSE, Cornelis, Roeland [NL/NL]; Delkant 6, NL-5311 CJ Gameren (NL). YKEMA, Durk [NL/NL]; Hemelvuur 13, NL-3454 SP De Meern (NL).  <b>(74) Agent:</b> OTTEVANGERS, S., U.; Vereenigde, Nieuwe Parklaan 97, NL-2587 BN The Hague (NL).		<b>(81) Designated States:</b> AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CR, CU, CZ, DE, DK, DM, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SL, SZ, TZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG).  <b>Published</b> <i>With international search report.</i>
<b>(54) Title:</b> STAR SHAPED ALUMINA EXTRUDATES AND CATALYST BASED THEREON  <b>(57) Abstract</b> <p>This invention is directed to star shaped alumina extrudates with a pore volume in the pores of a diameter over 1000 nm, as determined by mercury porosity, of at least 0.05 ml/g and a total pore volume between 0.5-0.75 ml/g. The extrudates have a length of between 2-8 mm, a length to diameter ratio of between 1-3, a side crushing strength of at least 50 N and a bulk crushing strength of at least 1 MPa.</p>		

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**FOR THE PURPOSES OF INFORMATION ONLY**

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

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DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

Title: STAR SHAPED ALUMINA EXTRUDATES AND CATALYST BASED THEREON

The invention is directed to transition alumina extrudates, suitable as catalyst, or as catalyst support, and the use of such extrudates in chemical reactions.

In catalysis alumina plays an important role, both as  
5 a catalyst support and as catalytically active material. As is mentioned in Kirk-Othmer, Third Ed, Vol. 2, pages 230-232, alumina can be used as catalyst in a Claus process, for dehydration of alcohols, such as the production of olefins from alcohol, and the reverse reaction, but also for the  
10 isomerisation of olefins. As interacting catalyst support alumina may play a role in hydrotreating catalysts, e.g. in cobalt or nickel-molybdenum oxides on alumina.

As a support alumina is frequently used for precious metal catalyst, such as in exhaust catalysts or for  
15 (de)hydrogenation reactions. As support for a nickel catalyst it may be used in (de)hydrogenation reactions such as for fat and oils hydrogenation, for hydrogenation of fatty nitriles or of nitro aromatic compounds or for oligomerisation of olefins.

20 The structure of the support, i.e. the BET surface area, the pore size and the pore volume distribution, forms an important aspect of the alumina or alumina based catalyst. In view of activity and selectivity it would be highly desirable to have an alumina product that is on the one hand  
25 highly porous, i.e. having a large volume in large pores, and that has a good mechanical strength and stability. Unfortunately these are requirements that are difficult to reconcile with each other.

In fixed bed processes shaped bodies of alumina are  
30 frequently used. An important aspect therein is the shape dependency of the pressure drop. Tablets and extrudates are the materials commonly used in fixed bed applications. In

order to minimise pressure drop, the use of star shaped extrudates would be most suitable. However, star shaped bodies, extrudates, tend to be prone to attrition due to the presence of the 'points' of the star.

5           It is an object of the invention to reconcile these various requirements in the form of a transition alumina extrudate, having a carefully balanced set of properties. Further objects and advantages will become clear from the following description of the invention and the preferred  
10   embodiments thereof.

          The invention is based thereon that the inventors have now been able to provide a star shaped alumina extrudate, having on the one hand an optimal structure, as indicated above and on the other hand a good strength.

15           The invention is accordingly directed to star shaped alumina extrudates with a pore volume in pores of diameter of over 1000 nm, as determined by mercury porosimetry, of at least 0.05 ml/g, a side crushing strength of at least 50 N and a bulk crushing strength of at least 1 MPa.

20           Surprisingly, this set of properties can be made available in one material, thereby providing a material with which chemical reactions can be made much more efficient, resulting in higher activity and/or selectivity. Also the material of the invention, when used in fixed bed reactors,  
25   provides a decreased pressure drop compared to regular extrudates having a cylindrical shape.

          The BET surface area, as determined by single point adsorption using the BET equation (as e.g. described by G. Sandstede et.al., Chem. Ing. Tech. 32 (1960), 413), should be  
30   at least 10 m<sup>2</sup>/g of alumina. This coincides with the requirement of using a transition alumina, i.e. not an  $\alpha$  alumina. Suitable alumina's are the various transition alumina's including  $\gamma$ -alumina,  $\delta$ -alumina,  $\epsilon$ -alumina,  $\kappa$ -alumina,  $\zeta$ -alumina,  $\theta$ -alumina and  $\tau$ -alumina. These alumina's  
35   have a large BET-surface area, generally in the range of 25 up to more than 100 m<sup>2</sup>/g.

The pore volume is a further important requirement, whereby it is on the one hand important that the total pore volume, as determined by mercury intrusion is sufficiently high and on the other hand that the pore volume in pores of over 1000 nm forms a substantial portion of the total pore volume. In absolute terms the total pore volume should be at least 0.50 ml/g, whereas the ratio of the pore volume in pores of over 1000 nm to total pore volume should preferably be more than 0.04. An alumina having those properties has good properties in terms of reactant accessibility, which makes it very suitable for all kinds of catalytic reactions requiring good diffusion of reactants and products through the alumina, thereby eliminating diffusion limitation problems as much as possible.

The pore volume and pore size distribution are determined by mercury porosimetry measurements, as described by J. Rouquerol et al in Pure & Applied Chem., 66(8),1994, pages 1752-1753, using the Washburn equation.

As indicated above, the use of star shaped extrudates is important in terms of pressure drop in relation to accessibility of the internal surface of the alumina. This also plays a role in eliminating diffusion problems. Star shaped extrudates can be defined as objects having some kind of central part or core, with three or more triangularly shaped extensions on the circumference thereof. Most preferred are star shaped extrusions having five extensions, as this provides the optimal balance between strength, porosity, pressure drop and accessibility. Another advantageous property of the star shaped extrudates is the fact that the ratio of external surface area to volume is more advantageous than in the case of conventional cylindrical extrudates or tablets.

The ratio of the length of the extrudates to the diameter is preferably between 1 and 3, whereby as diameter the distance is meant between two parallel planes on either side of the extrudate.

Important aspects of the material of the invention are also the strength characteristics. As indicated above a side crushing strength of at least 50 N and a bulk crushing strength of at least 1 MPa are essential herein. These parameters form the basis for the suitability of the extrudates for use in large scale reactors, like in the petroleum industry. When the extrudates meet these requirements, they can be used in huge fixed bed reactors, that require very strong material. The side crushing strength and the bulk crushing strength is defined as follows:

The side crushing strength (SCS) of extrudates is defined as the pressure (in Newtons) at which extrudates of 4.5-5.00 mm length are crushed, when treated under pressure between two flat plates on a AIKOH, 9500 series tester.

The bulk crushing strength (BCS) of a catalyst is defined as the pressure (in Megapascals) at which 0.5% fines (i.e. particles less than 0.425 mm) are formed when treated under a piston in a tube. For that purpose, 17 ml of catalyst particles, presieved on a 0.425 mm sieve, are loaded in a cylindrical sample tube (diameter 27.3 mm), and 8 ml steel beads is loaded on top. The catalyst is subsequently treated at different (increasing) pressures for three minutes, after which the fines are recovered and their percentages is determined. This procedure is repeated until a level of 0.5 wt.% fines is reached.

Another aspect of the strength of the material is the attrition, i.e. the amount of material that may break off of the extrudates upon use. This attrition, determined in accordance with ASTM D4058-87, should preferably be less than 5 wt.%, more in particular less than 3 wt.%.

The alumina extrudates having the above properties can be prepared by mixing transition alumina powder with a suitable binder in the presence of a liquid, usually water or an aqueous solution of a mineral acid such as hydrochloric, sulfonic or nitric acid, to form a paste, followed by extruding of the paste in the required star form, using a

suitable die and cutting the extruded strands of material to the required length. Optionally after drying, the extrudates are calcined.

It is possible to use various types of binder materials, such as those based on silica or alumina. Examples are colloidal silica, waterglass, or clays. It is preferred to use an alumina based binder or a binder that is removed during calcination, while providing and maintaining the required strength. An example of a suitable binder system is an alumina binder that gels under acidic treatment, for example by using organic or inorganic acids. The amount of binder material used in the preparation of the paste that is to be extruded will vary depending on the type of material and the required strength. Generally it will not be in excess of 30 wt.% based on the dry weight of binder and alumina together.

The invention will now be elucidated on the basis of an example.

#### EXAMPLE

1.5kg of aluminium trihydrate, containing 65 wt.% of  $\text{Al}_2\text{O}_3$ , with an average particle size of 30-50  $\mu\text{m}$  is mixed with 0.4 kg of alumina binder. The powders are mixed extensively while slowly adding diluted, aqueous  $\text{HNO}_3$  in an amount of 2 wt.%, calculated on the weight of the total amount of alumina.

Thereby the alumina binder is peptised. Mixing is continued until a relatively dry product is obtained. the intermediate product is extruded using a one-screw extruder, equipped with a die having starshaped holes and a cutting device.

The extrudates obtained are dried at 105°C for 16 hours and subsequently calcined at 850°C for one hour. Attached are two figures with photographs of an extrudate shown from two different angles.

The final product has been analysed for its physical properties with the following result:

N<sub>2</sub>-BET surface area 106 m/g<sup>2</sup>

Total Hg pore volume 0.56 ml/g

5 Pore volume in pores over 1000 nm 0.07 ml/g

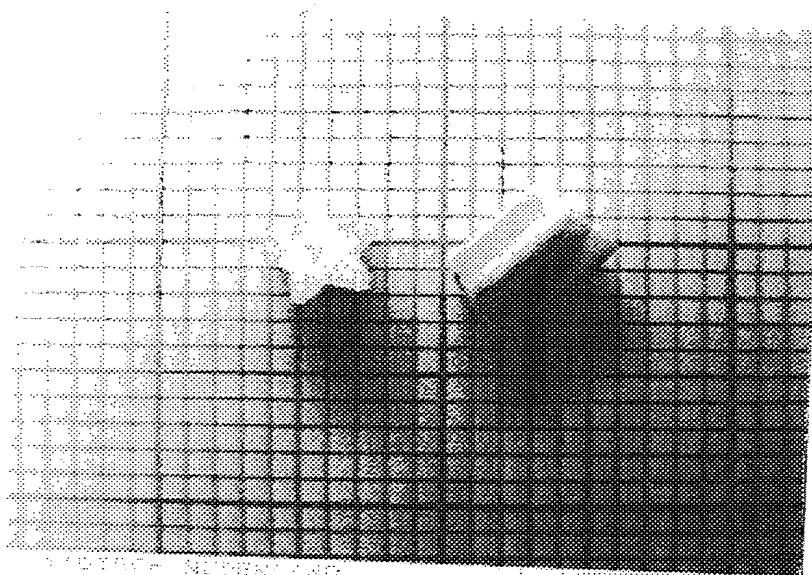
Side crushing strength 65 N

Bulk crushing strength 1.08 MPa



Claims

1. Star shaped alumina extrudates with a pore volume in pores of diameter of over 1000 nm, as determined by mercury porosimetry, of at least 0.05 ml/g, a side crushing strength of at least 50 N and a bulk crushing strength of at least 1 MPa.
2. Extrudates according to claim 1, having a length of between 2 and 8mm.
3. Extrudates according to claim 1 or 2, having a length to diameter ratio of between 1 and 3.
4. Extrudates according to claims 1-3, wherein the total pore volume a determined by mercury porosimetry is between 0.5 and 0.75 ml/g.
5. Extrudates according to claims 1-4, wherein the BET surface area is at least 75 m<sup>2</sup>/g.
6. Extrudates according to claims 1-5, wherein the attrition in accordance with ASTM D4058-87 is less than 5 wt.%, preferably less than 3 wt.%.
7. Catalyst, comprising at least one catalytically active material supported on an extrudate according to claims 1-6.
8. Catalyst according to claim 7, wherein the catalytically active material is selected from the group of metals, metal oxides, metal sulfides and combinations thereof.
9. Use of an extrudate according to claims 1-6 or a catalyst according to claim 7 or 8 in a chemical reaction.



# INTERNATIONAL SEARCH REPORT

International Application No

PCT/NL 99/00676

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 7 B01J35/02 B01J21/04

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)  
IPC 7 B01J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 008 424 A (BASF AG) 5 March 1980 (1980-03-05) claims 1,2 page 3, line 5 -page 5, line 7 example 1 figures 1,2 ---	1,4,5, 7-9
A	DE 33 15 105 A (LEUNA WERKE VEB) 17 November 1983 (1983-11-17) the whole document ---	1,4-9
A	WO 92 05870 A (MONSANTO CO) 16 April 1992 (1992-04-16) claims 1,3,4,10-12 page 16, line 34 -page 17, line 38 page 31; table 1 figures 5A,5B,11A,11B ---	1-3,7-9
	-/--	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

**Special categories of cited documents:**

- "A" document defining the general state of the art which is not considered to be of particular relevance
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Date of the actual completion of the international search

31 January 2000

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Name and mailing address of the ISA  
European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
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# INTERNATIONAL SEARCH REPORT

International Application No

PCT/NL 99/00676

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